The island of Syros consists largely of possibly-repeated sequences of glaucophane-bearing calcareous schists, mafic schists, dolomite marbles, and calcite marbles containing abundant aragonite pseudomorphs (Dixon, 1969; Hecht, 1984). Several discrete, fault-bounded packages of blueschist/eclogite-facies mafic rocks with minor serpentinite are also found on the island. Although the mafic rocks occur with a variety of textures and modes, most are either fine-grained, glaucophane-rich blueschists with a strong fabric or coarse-grained (>1cm), massive omphacite- or glaucophane-rich rocks. Based on textures and field relations, previous workers (e.g. Dixon and Ridley, 1987) have interpreted these rock types as meta-basalt and meta-gabbro, respectively. We have obtained 38 new whole-rock XRF and INAA analyses for 18 fine-grained and 20 coarse-grained samples. The fine-grained mafic rocks are chemically very similar and have basalt or basaltic andesite compositions compatible with an ocean floor or island arc origin. The coarse-grained mafic rocks vary more widely in composition and include samples that are significantly enriched or somewhat depleted in TiO₂, FeO, and V relative to the fine-grained mafic rocks. The chondrite-normalized REE patterns of the fine-grained mafic rocks are nearly flat with values in the range of 10 to 30. The REE patterns of 17 of the coarse-grained mafic rocks are depleted in LREE, have a clear positive Eu anomaly, and range in value from 5 to 20. We interpret these data to mean that the protoliths of the coarse-grained mafic rocks are indeed gabbros that have been chemically differentiated by fractional crystallization, whereas the protoliths of the fine-grained mafic rocks are largely undifferentiated ocean floor basalts. Our interpretation is consistent with the conclusions of previous workers based on field (Dixon, 1969), geochemical (Brocker, 1991; Seck et al., 1996), and isotopic (Putlitz et al., 2000) data. This result raises the interesting question of why a coarse-grained igneous protolith should lead to a coarse-grained metamorphic rock containing all new minerals. The massive character of the original gabbros appears to have had a strong influence on their metamorphism (coarse texture, little hydration) and deformation (little fabric, coherent blocks) during subduction and exhumation.